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Biodiesel production from neem towards feedstock diversification: Indian perspective

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ABSTRACT

In developing countries like India where 70% of country's petroleum needs are met by import, energy security assumes significance in view of uncertainty of supply and increasing price of petroleum fuels. Fuels of bio origin not only provide energy security, but also reduce emissions of harmful pollutants and greenhouse gases and ensure rural upliftment by increasing employment in agricultural sector. India cannot afford to produce biodiesel from edible oil seeds as it is done in the American and European countries. Extensive focus has been given on producing biodiesel from non-edible sources, specifically from Jatropha. Discrepancies between the expectation and realities regarding Jatropha as a feedstock necessitate efforts for diversification of the feedstocks. Scientific research should therefore be directed towards oilseeds like Karanja, Sal, Mahua, Neem, etc. that are widely available and sustainable to the diverse socio-economic and environmental conditions of rural India. Among them the evergreen neem with its wide availability and various useful uses may be a potential feedstock for biodiesel production. In this paper attempts have been made to overview the morphology of neem tree, various useful uses, physical and chemical characteristics of neem oil and optimized production process for biodiesel production from neem oil.

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1. Introduction

Energy is the key input for technological, industrial and socio-economical development of any country or region. Petroleum oil, natural gas, electricity and coal are the major commercial sources of energy across the world. Most of the current energy requirements in India are fulfilled by fossil fuels – coal, petroleum-based products and natural gas. In India, 95% of transportation energy is provided by oil and the demand for diesel is five times higher than the petrol demand. It was estimated that for sustaining India's 8% average annual economic growth and to support its growing population, India needs to generate 2- to 3-fold more energy than the present [1]. Since these commercial sources are finite and continuous depletion of fossil fuels is taking place, energy supplies in the world are becoming unsustainable. Dependence on fossil fuels also causes environmental problems both locally and globally. India ranks fifth after USA, China, Russia and Japan in producing high rate of emission and creating environmental and ecological imbalances.

Despite awareness drives and governmental initiatives towards popularizing and production of non-conventional energy, petroleum remains the primary energy source in India as a preferred fuel. Its consumption has been increasing at a very steep rate from 3.5 MT in 1950–1951 to 84.3 MT in 1997–1998 and projected to reach 200 MT in 2011–2012. The need of the hour is to conserve petroleum by its judicious use and to substitute it by other renewable resources wherever feasible. Renewable energy sources provide several significant benefits such as energy security, reduced emission of pollutants, greenhouse gases and increased employment in the agricultural sector. In its fourth assessment report, the intergovernmental panel on climate change (IPCC) confirmed that climate change was accelerating and if current trends continued, energy-related emissions of carbon dioxide (CO_2) and other greenhouse gases will rise inexorably, pushing up average global temperature by as much as 6 °C in the long term [2]. Hence, to reduce emissions of harmful pollutants and to conserve the environment, biofuels are going to play an extremely important role in developing countries like India by meeting country's energy demands.

Bio-diesel is one of the candidates as an alternate to diesel and is being explored world over because of its properties like high cetane number, low sulfur content and better lubricity than petro-diesel, which makes it an excellent fuel for diesel engines [3]. The various engine and vehicle tests conducted so far indicate the vehicle performance to be satisfactory with blends up to 20% of bio-diesel in diesel. The emissions of carbon monoxide and particulates are also lower with bio-diesel and petro-diesel blends [4]. Benefits for carbon credits exist on account of lower carbon dioxide emissions with use of bio-diesel. While blends up to 20% are quite common abroad, the Indian vehicle manufacturers are initially apprehensive and recommend use of only 5% or 10% of bio-diesel in diesel.

Biodiesel is produced from biological sources such as vegetable oils or animal fats using a biochemical process known as transesterification. The use of edible vegetable oils and animal fats for biodiesel production has recently been of great concern because they compete with food materials [5]. India is one of the leading importers of edible oils as demand exceeds the domestic production. As the demand for vegetable oils for food has increased tremendously in recent years, it is impossible to justify the use of these oils for fuel use purposes such as biodiesel production. Thus, the contribution of non-edible oils will be of great importance in the coming days as source for biodiesel production. It is estimated that the potential availability of such oils in India amounts to about 1 million tons per year, the most abundant oil sources are sal oil, mahua, neem oil and *Pongamia pinnata* [6].

In recent past, *Jatropha curcas* has been identified as major feedstock for biodiesel production in India because of its various speculated advantages such as wide adaptability, low requirement of water and fertilizers, pest resistance, low gestation period, high seed yield and oil content, easy propagation, and not browsed by cattle. But in practice, it shows wide variation in seed yield and oil content under different soil and agro-climatic conditions which present substantial risk and challenges that need to be addressed before huge monoculture plantation of this particular crop. Hence diversification of non-edible feedstocks is the call of the day towards ensuring sustainable supply of the renewable oil. This paper reviews the potential non-edible oil sources for production of biodiesel and focuses specifically on neem as a non-edible feedstock with an integrated approach for sustainability in renewable oil supply.

2. Energy security

2.1. India's energy outlook

India currently ranks as the world's 11th greatest energy producer, accounting for about 2.4% of the world's total annual energy production, while it ranks as the world's sixth greatest energy consumer, accounting for about 3.3% of the world's total annual energy consumption. India is one of the countries where the present level of energy consumption, by world standards, is very low. India, with over a billion people, today only produces 660 billion KWh of electricity and over 600 million Indians, a population equal to the combined population of USA and EU, have no access to electricity, and limited access to other clean, modern fuels such as LPG and kerosene [7]. Per capita energy consumption in India is less than 500 kgoe (Kilogram Oil Equivalent), compared to the global average of nearly 1800 kgoe.

It is estimated that India has only 0.4% of the world's proven reserves of crude oil. India meets about 70% of its petroleum requirements through imports which are expected to expand in coming years. During the last three decades, the production of crude oil in the country has increased from 6.82 million tons in 1970–1971 to 34.12 million tons in 2007–2008 [8]. In India, the

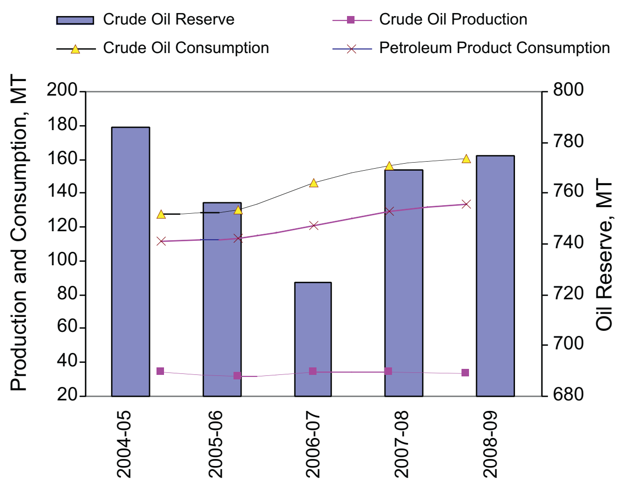

Fig. 1. Trend of crude oil production and consumption with respect to oil reserve.

volume of crude oil imported increased 9-fold from 11.66 million tons during 1970–1971 to 121 million tons by 2007–2008. During last 7 year, India's foreign exchange outflow due to this purpose has increased 5-fold because of the escalation of international oil prices [8]. The demand for diesel is projected to increase at an annual rate of 5.8% to 65 million tons in 2030. Petroleum fuel provides energy for 95% of transportation needs in India and the demand for diesel is 5-fold higher than the demand for petrol. India's share of crude oil production is about 1% of total world crude oil production while in consumption; its share is about 3.1% of total world consumption [9]. With a declining oil reserve, the quantity of crude oil and petroleum product consumption is increasing steadily. As shown in Fig. 1, there has been an increase of 26% in the consumption of crude oil and an increase of about 20% in the consumption of petroleum products during the last 5 years [8].

The World Energy Outlook has forecasted that by 2030, 94% of India's crude oil demand would be met by imports [9]. That means in near future India has to import a heavy amount of crude oil for meeting country's energy demand and consequently pay a huge foreign exchange for it. The country's energy demand is expected to grow at an annual rate of 4.8% over the next couple of decades. So for ensuring energy security it has become necessary to tap all possible energy options, mainly renewable and diversify the energy basket. Biofuels are going to play an extremely important role in meeting India's energy needs. To keep pace with its rapidly expanding economy and energy demand with environmental sustainability, Indian Government has been setting targets and developing strategies, policies, and investment plans in biofuels to enhance energy security and explore alternative energy sources. The recent increase of oil prices, energy security fears, increasing foreign exchange outflow due to massive petroleum import and international environmental regulations give cause for a more serious consideration of renewable energy in India.

2.2. Potential of renewable energy

Though the installed capacity is not that appreciable, India has huge potentials for the effective use of renewable energy (Table 1). India is the world's fourth largest producer of wind power after Denmark, Germany, and Spain. Other renewable energy technologies, including biomass, wind, solar, small hydro (less than 25 MW), and waste to energy are also growing. India's renewable energy sources in the perspective of potential and installed capacity can be categorized as centralized and decentralized systems. Centralized systems can be of two types – grid interactive renewable power and off-grid or distributed renewable power. Today, the installed capacity of the grid-interacted renewable power is about 16,000 MW [10]. Family size biogas plants cover most of the decentralized renewable energy system. With a potential of 12 million biogas plants, India has nearly 4.2 million installed biogas plants [10].

The Electricity Act, 2003, requires State Electricity Regulatory Commissions to specify a percentage of electricity that the electricity distribution companies must procure from renewable sources [7]. This has contributed to acceleration in renewable-electricity capacity addition, and over the past 3 years, about 2000 MW of renewable-electricity capacity has been added in India every year, bringing the total installed centralized grid-interactive renewable capacity to over 16,000 MW. Of this, about 11,000 MW is based on wind power; India now has the fourth largest installed wind capacity in the world. There has been an overall 6-fold increase in the installed capacity of grid-interactive renewable power in India over the last decade with about 10-fold increase in the wind power sector contributing about 70% of the total renewable power capacity. The installed capacity of off-grid renewable energy is close to 55 MW with the largest contribution from biomass and cogeneration system (about 40 MW)[10].

Biomass is the major source of energy in rural India; 90% of the rural population use biomass that consists of crop residue, animal wastes, fuel wood and cow dung, etc. as domestic fuel for cooking and other purposes. India has huge potential for first generation biofuels that are produced from biomass consisting of vegetable oils, animal fats, and biodegradable wastes from agriculture and industry using conventional technologies. In recent times much focus has been on biofuel production that included mostly ethanol and biodiesel. Though ethanol industry in India has already grown up, the biodiesel industry is required to be given more attention.

2.3. Initiatives on biodiesel in India

To encourage production of bio-diesel in the country, the Ministry of Petroleum and Natural Gas announced a Bio-diesel Purchase Policy, in October 2005, which became effective from 1.1.2006. The Policy has identified 20 purchase centres of the public sector Oil Marketing Companies (OMCs) all over the country. The OMCs will purchase bio-diesel meeting the standards prescribed by the Bureau of Indian Standards (BIS), from those bio-diesel manufacturers who register with them after satisfying the technical specifications, at a specified delivered price. Depending on demand and preparedness, the OMCs could also open more purchase centres.

In India, biodiesel research, production, and marketing are in the early stages of development. *Jatropha curcas* has been identified for India as the most suitable tree borne oilseed (TBO) for production of biodiesel both in view of the non-edible oil available from it and its presence throughout the country. Presently, in some Indian villages, farmers are extracting oil from Jatropha and after settling and decanting it they are mixing the filtered oil with diesel fuel. In India, a biofuel mission was started in 2003 and the Government has announced a biofuel policy in September 2008. The Indian Government approved a national policy for setting up an empowered national biofuel coordination committee and a biofuel steering committee [12]. Draft policy envisages the following:

1. A target of blending ethanol and biodiesel with petrol and diesel at the rate of 20% by 2017.
2. Community-based biodiesel production from non-edible oils in waste, degraded, or marginal lands.
3. Minimum support price for biodiesel and minimum purchase price for bioethanol.
4. Biodiesel and ethanol may be brought under ambit of "Declared Goods" to ensure unrestricted movement.

Table 1
Details of estimated potential and installed capacity of renewable power [10,11].

Renewable power	Estimated potential (MW)	Installed capacity (MW) as of		Increase in 10 years
		1999–2000	2009–10	
Centralized				
Grid-interactive				
1. Biomass/cogeneration bagasse	21,881	171	2140	11.50-fold
2. Wind	45,195	1022	10,900	9.7-fold
3. Small hydro	15,000	1218	2560	1.1-fold
4. Waste to energy (urban)	2700	2	65	31.5-fold
5. Solar	50 MW/Sq km	1	9	8-fold
Total		2414	15,694	5.5-fold
Decentralized				
Family size biogas plants (nos)	12 million	2.9	4.1	

5. No taxes and duties to be levied on biodiesel.

The National Biodiesel Mission was implemented in two stages. The first was a demonstration project that was carried out over the period 2003–2007 for cultivating 400,000 ha of Jatropha. Reputed Indian automobile companies and their research centres have been actively engaged with research on various aspects of production and utilization of biodiesel. Indian Oil Corporation (IOC) has been at the forefront of technology development for production of biodiesel from various edible and non-edible oils and its application in vehicles. IOC has taken initiative to start commercial pilot projects on biodiesel production in states, which have clear land allotment policy favorable to commercial energy-crop plantation. Research and development Centre of Indian oil Corporation was the first in the country to undertake this programme in a scientific manner and systematic way. Oil from *Jatropha curcas* and *Pongamia pinnatta* were evaluated and analysed for various components. After hydrolysis, the fatty acids present in these two oils were compared with those of soybean, rapeseed, palm, etc. The overall resemblance of these fatty acids indicated the pre-feasibility of using these as feedstock for bio-diesel production.

Besides the automobile industries, educational Institutes also have been conducting research on biodiesel production and process optimization with various feedstocks. IIT Delhi developed transesterification process for Jatropha and Karanja oil. A set of process conditions for transesterification was optimized for production of JOME and KOME. The physico-chemical properties of the Jatropha oil, Karanja oil, neat petro-diesel, neat bio-diesel (B100) and its blends of 5% (B5), 10% (B10) and 20% (B20) were also determined at the IIT, Delhi as per the ASTM standard methods [13].

Punjab Agricultural University, Ludhiana conducted studies on bulk production and use of selected plant oil esters as alternate renewable fuel for diesel engine. Feedstocks considered included Linseed oil, sunflower oil and rice bran oil. After optimization of various parameters for biodiesel production, CIAE, Bhopal developed an aqueous ethanol (160 proof) and catalyst (KOH) based batch biodiesel production unit of 5–8 l/d utilizing glassware. Its performance was evaluated for production of Karanj and Jatropha bio-diesel and on an average recovery of bio-diesel of 83–88% was observed against that 90–93% from methanol process [13]. CSIR and Daimler Chrysler have jointly undertaken a successful 5000 km trial run of Mercedes cars using bio-diesel as fuel.

As a part of Indian Railway's initiative, Shatabadi Express was run on 5% blend of bio-diesel. Field trials of 10% bio-diesel blend were also done on Lucknow–Allahabad Jan Shatabdi Express also through association with IOC. NOVOD has initiated test run by blending 10% bio-diesel in collaboration with IIT, Delhi on Tata Sumo and Swaraj Mazda vehicles. Exhaustive emission testing was carried out at R&D centre of IOC on diesel cars and buses of Haryana Roadways. Using 10% and 20% blends of bio-diesel in diesel, there was 20% and 25% reduction in CO emission and 5% and 10% reduction in particulate matter, respectively, on a diesel car.

3. Diversification of biodiesel feedstock

3.1. Non-edible oils: renewable oils for India

The choice of feedstock for biodiesel production is country specific and depends on availability. The selection of vegetable oil or renewable oil that is used for transesterification for producing biodiesel varies from region to region depending on the social, environmental economical and agro-ecological parameters. The United States uses soybean, Europe rapeseed and sunflower, Canada canola, Japan animal fat and Malaysia palm oil. Edible oils like soybean, sunflower, rapeseed and palm are used as main biodiesel feedstocks throughout the world. Oil from rapeseed has been the great choice in the early days and is still leading with a share of over 80% as a raw material source with highly suitable properties; sunflower oil takes second place with over 10%, followed by soybean oil [14].

Oilseeds and edible oils are two of the most sensitive essential commodities in India. In spite of the production of large volume of oils with diverse species, India is not self-sufficient in edible oils. During the last two decades, the edible oil consumption has increased at a compound average growth rate of 4.25% from mere 4.959 million tons in 1986–1987 to 12.191 million tons in 2007–2008 [15]. The edible oil consumption in the country is presently growing and likely to remain heavily dependent on imports. It is predicted that by 2015, the demand for edible oil in India would be 20 million tons per annum [16]. Huge demand for domestic consumption, stagnant production growth, and foreign exchange outflow make edible oil an unviable option for biodiesel in India. Therefore, the Indian government decided to explore non-edible oil sources as alternate feedstocks for biodiesel production in India.

Non-edible oil is most suitable feedstock for biodiesel since the demand for edible oil exceeds the domestic supply. Non-edible oil is most suitable feedstock for biodiesel since the demand for edible oil exceeds the domestic supply. India has a vast untapped potential of nonedible oilbearing plant species distributed throughout the country: 300 species of trees have been reported to produce oil bearing seeds [17]. Non-edible oils from the sources such as neem, mahua, pongamia, karanji, babassu, and jatropha, are easily available in many parts of the world including India, and are very cheap compared to edible oils [18]. In India, there are several non-edible oils from different species such as Pungam (*Pongamia pinnata*), Jatropha (*Jatropha curcas*), Neem (*Azadirachta indica*), Mahua (*Madhuca indica*) and Simarouba (*Simarouba indica*), which could be utilized for biodiesel production processes [19]. According to a survey conducted in 2002, 12 species have been selected for its

Table 2
Potential of tree born non-edible oils in India [21–23].

Sl. no.	Tree born oil sources	Botanical name	Oil content (%)	Potential seed and oil yield (million tons/year)	
				Seed	Oil
1	Karanja	*Pongamic pinnata*	27–39	0.02	0.055
2	Jatropha	*Jathropha Curcas*	30–40	0.05	0.015
3	Kusum	*Scheleichera oleosa*	28–34	0.08	0.025
4	Neem	*Azadirechta indica*	30	0.50	0.100
5	Sal	*Shorea robusta*	12–13	1.50	0.180
6	Mahua	*Madhuca indica*	35–42	0.50	0.180
7	Mango	*Mangifera indica*	7.5	0.50	0.045
8	Tumba	*Citrulluscollocynthis*		–	0.017
9	Kokum	*Garcinia indica*	25–34	–	0.0005
10	Jojoba	*Simmondsia chineaca*	45–50	–	–
11	Chullu	*Prunus armeniaca*	45–50	–	0.0001
12	Nahar	*Mesua ferrea L.*	60–70	–	0.01
13	Pilu	*Salvadora oleoides*	38–42	–	0.017
14	Rice bran	*Oryza sativa*	15–23	–	0.474
15	Phulware	Cheura	60–62	–	0.003

importance of present industrial usage and abundance in distribution [20]. It is estimated that (Table 2) the potential availability of such oils in India amounts to about 1 million tons per year [6]; the most abundant oil sources are sal oil (180,000 tons), mahua (180,000 tons), neem oil (100,000 tons) and *Pongamia pinnata*, also known as Karanja oil (55,000 tons).

There has been substantial research on Jatropha, Pongamia, Karanja and Mahua for their suitability in Indian conditions and also to meet the automobile blending requirements. However, efforts need to be focused on neem which has been considered as highly worth full tree for its multi purpose uses. The major bio-chemical component that is extracted from neem oil for its medicinal purpose is 'Azardiratchi'. After extracting this important component, the remaining part of the oil can be transesterified for biodiesel production that can be considered as a noble approach for its integrated application.

3.2. *Myths and facts about Jatropha*

Jatropha is a wild plant with large variations in growth, production and characteristics. Contrary to popular belief, Jatropha needs optimal quantity of fertilizer and irrigation for increased seed and oil yield. Seed yield was found to depend on various factors such as rainfall, soil type soil fertility, genetics, plant age and propagation method, spacing, pruning, fertilizing, and irrigation [24]. A wide range of variation in Jatropha seed yield (0.4–2 tons/ha/year) has been reported in the literature. Jatropha is susceptible to various pests and diseases. An outbreak of pests and diseases could wipe out total plantations in one stroke. So various issues regarding crop management, water use efficiency, drought resistance, yield, pest management, land suitability should be addressed before complete dependence on Jatropha. Even though Indian search/research on Jatropha has been more than a century old, co-coordinated research is needed to attain improved yield [25]. Major limitations of Jatropha are discussed below.

3.2.1. *Wide variation in yield and oil content*

The seed yield and oil content of the feedstocks are the major determining factors for the economic viability of biodiesel production. Jatropha exhibits great variability in productivity between individuals. Recently Kaushik et al. [26] recorded coefficients of variance between 24 provenances of Haryana state, India, which indicate a dominant role of environment over genetics in seed size, seed weight and oil content. They reported a wide variation in oil content (21–40%). Table 3 reflects the variation of seed and oil yield from Jatropha that necessitated a re-look on the feedstock.

3.2.2. *Irrigation and fertilizer requirement*

When grown without irrigation, Jatropha gives one fruiting cycle per year, but if abundant water is made available to the tree on a year-round basis, up to three fruiting cycles may occur within a year. Jatropha has shown high demands for nitrogen and phosphorous [31]. On degraded sites JCL plants are found to respond better to organic manure than to mineral fertilizers [32]. Hence, satisfactory yield from waste land planting without proper care cannot be expected.

3.2.3. *Pests and diseases*

It has been a common belief that Jatropha is not prone to pests and diseases in such extent to cause economic damage. However, in continuous Jatropha monocultures in India economic damage has already been observed [33]. The major problems in Jatropha cultivation are caused by the scutellarid bug Scutelleranobilis and the inflorescence and capsule-borer Pempeliamorosalis [33]. Requirement of regular irrigation and fertilizer application is expected to enhance the pests and disease infestations in commercial monocultures [34].

Jatropha has several other drawbacks. For example, its leaves are not suitable to be used as fodder for livestocks. This means that plantation of Jatropha on grazing lands is likely to worsen the food crisis. Jatropha yields insignificant amount of wood per tree. The wood density of Jatropha is very low, ranges from 0.22 to 0.37 [35]. Even though a higher biomass accumulation rate of 5 tons/ha/year was estimated, the net carbon sequestration rate is only 1.05 tons C/ha/year ($5 \times 0.3 \times 0.7$) [36].

Hence, it is time to look for other alternatives such as Pongamia, Mahua, Neem, and Seemaruba. Among them neem has not yet been commercially tried and more research has to be done in this direction. Billions of neem trees exist all over India. If oil is extracted from the seeds at village level by traditional expellers, few million tons of oil will be available for lighting the Lamps in rural area. However, in spite of good demand, only about 25–30% of the neem seed is collected in India, indicating it as a large untapped potential.

Table 3
Variation in yield of Jatropha [27–30].

Type of data	Reported yield on maturity (tons/ha)
Primary data from block plantations	1.0–1.2
Estimates for poor soil (Kutch)	1.6–2.5
Estimates for average soil	3.3–5.0
Estimates for poor soil with low nutrient content	1.5–2.0
Estimates for rain-fed and irrigated conditions	3.0–5.0

4. Neem and its value chain

4.1. Morphology

The evergreen neem (*Azadiractha indica* A. Juss.) tree belongs to the mahogony family and is native to India and Burma. Today, the neem is well recognized in at least 30 countries world-wide, in Asia, Africa and Central and South America. Its life is more than 200 years and grows everywhere, from semi-arid and sub-humid conditions up to 1500 above sea level [37]. It is found in tropical and subtropical climates and survives at annual mean temperature 21–32 °C. It can grow where rainfall is below 400 mm and also tolerates temperature as high as 48 °C. Neem can tolerate extremely dry condition as well as sub humid condition. It cannot survive under freezing conditions, particularly in the early stages of growth. Neem can be grown on very poor soils that may be very stony, dry and have hard calcareous or clay pan even at a very shallow depth. Neem grows on almost all types of soils including clay, saline and alkaline soils, having pH up to 8.5. It can be grown directly by sowing its seed or by transplanting nursery raised seedlings with little care. It is not palatable by livestocks and is also less affected by diseases and pests.

4.2. Multipurpose uses of neem tree

Neem is a golden tree that has gained world-wide importance owing to its multiple uses. Besides agroforestry, it is used in pest control, toiletries, cosmetics, pharmaceuticals, plant and animal nutrition and energy generation. Neem trees are considered to be a divine tree in India because of their numerous valuable uses. The commercial value of neem has been known since the Vedic times (about 1500 B.C.). Every part of neem tree viz., leaf, flower, fruit, seed, kernel, seed oil, bark, wood, twig, root, etc. has been in use and traded in various purposes [38].

4.2.1. Neem in sustainable agriculture

The use of neem tree may play an important role for sustainable development when used in an integrated approach. Contributions of neem towards sustainable development include better pest and nutrient management, human health, and environmental conservation. Pesticides made from neem that neutralise almost 500 pests worldwide, are much safer and environment friendly than synthetic pesticides. Chemical insecticides work on the digestive or nervous system of pests. But neem compounds work on the insect's hormonal system and thus restricts the development of resistance in future generations. Neem extracts alter the life-processing behavior in such a way that the insect can no longer feed, breed or undergo metamorphosis.

With its multiple applications including that in organic farming, it plays an effective role in sustainable agriculture. The leftover seed cake after oil extraction is used as an excellent organic fertilizer and is several times rich in plant nutrients than manure [39]. It is rich in nitrogen (2–3%), phosphorous (1.0%) and potash (1.4%) [40]. The effect and economics of using neem cake, with one or more than one fertilizer like urea, superphosphate, etc. have been studied and found that there is a saving of 25–50% urea nitrogen coupled with improved yield [41].

4.2.2. Environmental service

Neem is believed to purify the air and environment of harmful materials. It is believed that the shade of the neem tree not only provides cool but it also prevents the occurrence of many harmful diseases. During hot summer season the temperature under the neem tree is about 10 °C lesser than the surrounding temperature. It is one of the very few shade-giving trees that survive in the drought prone areas. Thats why it is believed to be a life giving tree for the dry coastal regions of India.

4.2.3. Neem for reforestation and agro-forestry

Neem is a unique tree for agroforestry as wind breaks and as single scattered trees. It is compatible with many crops as its deep root system avoids competition for soil moisture and nutrients with agricultural crops [37]. Neem plantations in rows of 4–6 m apart allow satisfactory growth and yield of various arable crops. Neem can also be used for afforestation in saline soils using soil replacement technique and will make the site for moderate pastured [39]. Being a hardy, multipurpose tree, it is perfect for reforestation programs and for rehabilitating degraded, semiarid and arid lands. Neem trees have been used successfully as shelterbelts in arid and semi-arid regions of Sahel, Sudan and Nigeria [42]. Since neem wood is very thick that belonging to the mahogany family of trees, it may be used for heating, construction, furniture and craft-making.

4.2.4. Medicine from neem

All parts of the tree (seeds, leaves, flowers and bark) are used for preparing many different types of medicines. Neem is used for preparing traditional Ayurvedic medicines for the treatment of fever, leprosy, malaria, ophthalmia and tuberculosis. Neem is also known to possess curative and preventive properties against various skin infections and other problems. Extracts of neem leaves are helpful for malaria, diabetes, scrofula, erysipelas, etc.

5. Neem oil: an alternate feedstock for biodiesel

A neem tree can produce many thousands of flowers, and in one flowering cycle, a mature tree may produce a large number of seeds. Neem trees start bearing harvestable seeds within 3–5 years, and full production may be reached in 10 years, and this will continue up to 150–200 years of age. A mature neem tree may produce 30–50 kg of fruit each year. By rough estimate India nearly has 20 million neem trees. Indian neem trees have a potential to provide 1 million tons of fruits per year and 0.1 million tons of kernels per year (assuming 10% kernel yield). Neem seeds yield 40–60% oil [39]. Considering the conservative level of oil content of only 30%, annual neem oil production in India could be to the tone of 30,000 tons. With an integrated approach, the produce will lead to azadirachtin production (assuming 0.2–0.5% of kernel by weight) of about 200–500 tons per year.

5.1. Methods of oil extraction

The two principal methods used to obtain oils from neem seeds are: expulsion, where the oil is obtained through pressing (crushing) of the seed kernel through cold pressure or through a process incorporating temperature controls, and extraction where the oil is removed from the seeds using a solvent. Some of the active components of neem oil are susceptible to heat, so cold pressed oil is preferred. The composition and the properties of the oil obtained by these two methods are quite different because these methods are unable to remove the same mix of components in the same proportion. Extracts expelled from the seed will also contain water expelled from the seed by the same process and the aqueous material will carry along with it liminoids such as azadirachtin which has high pesticidal activity (US Patent 5503837 – Co-extraction of azadirachtin and neem oil).

Studies are going on to extract the hydrophilic azadiractin portion of the neem seeds and hydrophobic neem oil portion of the seeds in a single extraction step. However, the solvent extracted neem oil is of lower quality as compared to cold pressed neem oil. Liauw et al. [43] extracted neem oil using two solvents ethanol and *n*-hexane at five different temperatures (30, 35, 40, 45 and 50 °C)

and studied the properties of the oil extensively. The maximum yield of 44.29% and 41.11% were obtained at 50 °C using *n*-hexane and ethanol, respectively. They also reported that the quality of neem oil decreased as the extraction temperature increased.

5.2. Properties of neem oil and neem biodiesel

Neem oil is generally light to dark brown in colour, bitter in taste and has a strong odour. It mainly consists of triglycerides and large amount of triterpenoids which are responsible for bitter taste. Neem oil also contains steroids like campesterol, beta-sitosterol, stigmasterol and a plethora of triterpenoids of which azadirachtin is the most well known. The azadirachtin content of neem oil varies from 300 ppm to over 2000 ppm depending on the quality of the neem seeds crushed. After separating azadirachtin which is used as a great insect repellant and is a systemic pesticide, most part of the neem oil could well be used for producing biodiesel. Among the fatty acid profiles of seed oils of 75 plant species having 30% or more fixed oil in their seed/kernel, neem (*Azadirachta indica*) oil is one of the most suitable oil for use as biodiesel [44].

5.2.1. Fatty acid profile of neem oil

The fatty acid profile can crudely be determined by wet chemical methods which include the iodine and saponification values [45]. The iodine value indicates the total amount of unsaturated fatty compounds in a sample, but it does not give any information on the nature of the unsaturated compounds. It also does not provide any information regarding saturated compounds. The saponification value is linked to the average molecular weight of the sample of fatty compounds. The wet chemical methods are time-consuming and hence are generally replaced by more informative chromatographic and spectroscopic methods.

Chromatographic methods are used for qualitative and quantitative analysis of fatty acid methyl esters. The major chromatographic methods are gas chromatography (GC) and liquid chromatography (LC). Chromatographic method separates different compounds based on their physical properties.

In GC, the separation is mainly done by the differences between boiling points and structures (polarity) of the individual compounds. The sample in low concentration is usually dissolved in an organic solvent and then injected into the gas chromatograph. After injection, the sample is separated on a column which is a long, thin capillary tube and contains materials with which the sample components react more or less strongly depending on their structures (polarity). There is a detector to detect the material eluting from the column at a certain retention time and this time will be recorded and shown in the chromatogram. Generally, the integrated value of the peak and amplitude over time is proportional to the amount of material causing them. This constitutes the usefulness of GC in quantifying the amounts of components in a mixture [45]. Standards are also used in GC which helps in knowing the nature of specific components in a mixture.

In LC, the separation is done based on the differences in solubility of the components in a solvent. The separation is performed in a column like GC. The LC analysis is generally carried out at room temperature.

Chromatographic methods only detect if a compound is eluting, not its identity or structure. The identity or structure needs to be established through the use of standards as far as possible. Spectroscopic method of detection in combination with a chromatographic method yields more detailed information. The most common method in combination with chromatography is mass spectrometry (MS) where the spectra in MS record how a compound is broken up into fragments by means of energy (usually a beam of electrons). The way a compound splits into fragments is characteristic of its structure [45].

Table 4
Fatty acid profile of neem oil [14,18,23,44].

Fatty acid	Range	Mean	Standard deviation
Oleic	25–62	45.75	6.869498
Linoleic	2.3–17.9	13.68	4.344633
Palmitic	13.6–33	17.78	4.283148
Stearic	9.0–24.0	17.70	2.532456
Linolenic	0.4–1.3	0.83	0.450925

The amount and type of FA present in the feedstock oil determines the various physical and chemical properties of the produced biodiesel. The fatty acid profile of neem oil is given in Table 4. The percentage basis representation of the average values of the major fatty acids are also shown in Fig. 2.

5.2.2. Neem biodiesel

Neem oil can be used as fuel in diesel engines directly and by blending it with methanol. Neem oil with a high calorific value matches diesel. Its blends with diesel substituting nearly 35% of the later have been suggested for use without any major engine modification and without any worthwhile drop in engine efficiency [46]. Engine tests with neem oil and neem biodiesel were done in India and Bangladesh, showing satisfactory engine performance [47,48]. Yield of biodiesel from different non-edible oils (*Jatropha curcas*, *Pongamia pinnata*, Madhuca Indica and Azadirachta Indica) which are commonly available in India were examined and the results recommended the biodiesel production from Azadirachta Indica oil on the basis of high yield and quality of biodiesel [49]. The economic evaluation has also shown that the biodiesel production from neem is very profitable [49]. The study conducted by Sekhar et al. [50] supports the production of biodiesel from non-edible neem oil as a viable alternative to the diesel fuel. SathyaSelvabala et al. [51] esterified free fatty acids present in *Azadirachta indica* (neem) oil with synthesized phosphoric acid modified catalyst. During the esterification, the acid value was reduced from 24.4 to 1.8 mg KOH/g oil.

Physical and chemical properties of neem oil, neem methyl ester and conventional diesel are presented in Table 5. The fuel properties of neem biodiesel were within the limits and comparable with the conventional diesel. Except calorific value, all other fuel properties of neem biodiesel were found to be higher as compared to diesel [50].

5.3. Transesterification – biodiesel production process

The most commonly used method to produce biodiesel from crude vegetable oils and animal fats are to transesterify vegetable oils and animal fats to an alkyl ester to reduce their viscosity and increase in their volatility [53]. The reaction of fats or oils with alcohols to produce biodiesel is called transesterification. In general, there are two methods of transesterification. One method employs

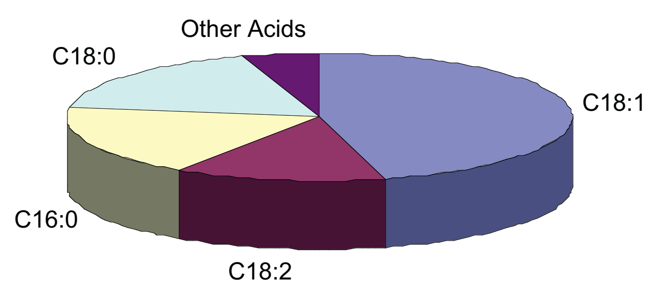

Fig. 2. Fatty acid composition (%) of neem oil. C16:0 = palmitic acid; C18:0 = strearic acid; C18:1 = oleic acid; C18:2 = linoleic acid. Other acids including mainly linolenic acid.

Table 5
Properties of neem oils and its ester [23,47,48,50,52].

Properties	Diesel	Neem oil	Neem biodiesel
Density (kg m^{-3})	830	912–965	820–940
Viscosity (cSt)	4.7	20.5–48.5	3.2–10.7
Flash point (°C)	60	214	120
Cetane number	45	31–51	48–53
Calorific value (MJ kg^{-1})	42	32–40	39.6–40.2
Sulfur (ppm)	0.042	1990	473.8
Saponification value	–	175–200	–
Iodine value	–	65–80	–
Titre (°C)	–	35–36	–
Fire point (°C)	65	222	128
Pour point (°C)	−16	10	2
Cloud point (°C)	−12	19	9
Total glycerin (%)	–	–	0.26
Free glycerin (%)	–	–	0.02
Oxidation stability (h), 110 °C	3–6 min	12.4	7.1
Cold filter plugging point (°C)	–	11	–
Carbon residue (% mass)	0.17	–	0.105
Water content (%)	0.02	0.098	0.036

a catalyst and another one is non-catalyst options such as supercritical processes, and co-solvent systems. A catalyst is employed to increase the reaction rate and yield.

Various catalysts used are base catalysts that include NaOH, KOH, and NaMeO, acid catalysts that include H_2SO_4, H_3PO_4, and $CaCO_3$ and lipase enzymes. Methanol and ethanol are the two main light alcohols used for transesterification process. For biodiesel production alkaline or acidic catalysts are frequently used. The alkali-catalyzed reaction is reported to be much faster than acid-catalyzed one [54]. Enzymes are also used as catalyst for transesterification. Glycerol is an important byproduct which can be burnt for heat or used as feedstock in cosmetic industry. Most of the current commercial biodiesel production is dominated by base catalyst process. Base catalyzed biodiesel production process generally consists of unit operations of transesterification reaction, distillation for recovery of excess alcohol, water washing for separating biodiesel from glycerol, catalyst and alcohol, distillation for crude biodiesel purification and glycerol purification. Fig. 3 shows the process flowchart of generalized biodiesel production methods.

5.4. *Catalysts for biodiesel production*

5.4.1. *Alkaline catalysts*

The alkali catalyzed transesterification process is affected by free fatty acid and water content of the feedstocks. For alkali catalyzed process the FFA content of the feedstock should be less than 1%, otherwise the soap produced by the reaction between FFA and alkali will lower the catalyst efficiency and consequently decrease the biodiesel yield. Most commonly used alkali catalysts are Potassium hydroxide, sodium hydroxide and sodium methoxide. Singh et al. [55] reported that methoxide catalysts give better biodiesel yield than hydroxide catalysts. But due to the high price and hygroscopic nature of methoxide catalysts, they are preferably used for large continuous flow biodiesel production process while hydroxide catalysts are used by small biodiesel producers.

5.4.2. *Acid catalysts*

Acid catalyzed transesterification process is much slower than alkali catalyzed process. It is preferred when FFA and water content in the feedstock are more. An acid catalyst reaction needs a larger amount of alcohol to triglyceride ratio than alkali catalyzed one for achieving the same biodiesel yield for a given reaction time. The commonly used acid catalysts are sulfuric, sulfonic and hydrochloric acids.

However, heterogeneous catalysts and biocatalysts are also becoming attractive in recent days. Acid and base catalysts are soluble in alcohol and so they pose problem in downstream separation of biodiesel. Gryglewicz [56] reported that use of heterogeneous catalyst can significantly simplify biodiesel separation process as they are insoluble in alcohol and thus reduce the production cost. A co-solvent such as tetrahydrofuran is added to the oil, alcohol, heterogeneous catalyst mixture to increase the mass transfer rates between the reactants. As a result the reaction completes within 5–10 min at a temperature of 30 °C with no residues of catalyst left either in the ester or in the glycerol phase [45]. When biocatalysts like immobilized lipases are used, it may be possible to extract the oil and transesterify it simultaneously. But enzyme catalyzed reactions requires much longer time than alkali catalyzed one and the processing cost is also higher. Hence, this has not yet been commercially tried.

The process selection for transesterification depends upon the FFA content of neem oil and FFA content varies accordingly with the storage period. Ragit et al. [47] used freshly extracted filtered neem oil having free fatty acid content of 0.403% for biodiesel production. They used single step base catalyzed transesterification as the FFA content of the oil was below 1%. Sekhar et al. [50] produced biodiesel from crude neem oil having 26% FFA content using two step processes – acid pretreatment followed by base catalyzed transesterification.

The FFA content in freshly extracted oil remains less. FFA content of oil increases due to long storage, oxidation and hydrolysis reaction. The higher percentage of FFA had significant effect on transesterification of glycerides. The higher FFA content would lead to soap formation. As a result the separation of biodiesel from glycerol would be more difficult and also the yield would be less. Hence, an alternate method such as pretreating the oil with concentrated sulfuric acid is used to reduce the FFA content of the oil below 1%. The base transesterification is then adopted to produce biodiesel.

5.5. *Optimization of transesterification*

The physical and chemical properties of the feedstock oils are the deciding factors which influence the modification of the input ratios of the alcohol, reagent and reaction catalyst as well as alterations to reaction temperature and time, in order to reach optimal bio-diesel production results. The most important variable that influence the transesterification reaction are reaction temperature, ratio of alcohol to vegetable oil, catalyst, mixing intensity and purity of reactants. Selection of available processes along with their parametric values for biodiesel production from neem oil is given in Table 6.

Generally the transesterification reaction is conducted below the boiling point of the alcohol used, 60 °C for methanol and 78 °C for ethanol. Higher reaction temperature can cause increase in the reaction rate and decrease in the reaction time. A 3:1 molar ratio of alcohol to triglyceride is stoichiometrically needed for the completion of the reaction. But in practice a molar ratio as high as 6:1 is needed to drive the equilibrium to maximum ester yield. The biodiesel yield increases with the increase in reaction time. Base catalyzed transesterification reactions are usually completed within an hour. Vicente et al. [57] reported that an increased amount of alkali catalyst increased the amount of soaps produced through triglyceride saponification and as a result the biodiesel yield decreased. So for getting maximum biodiesel yield optimized combination of all the reaction variables should be considered.

5.6. *Application of neem biodiesel to CI engine*

Several studies found that brake thermal efficiency (BTE) for methyl esters of neem oil is nearly close to diesel [47,48,58].

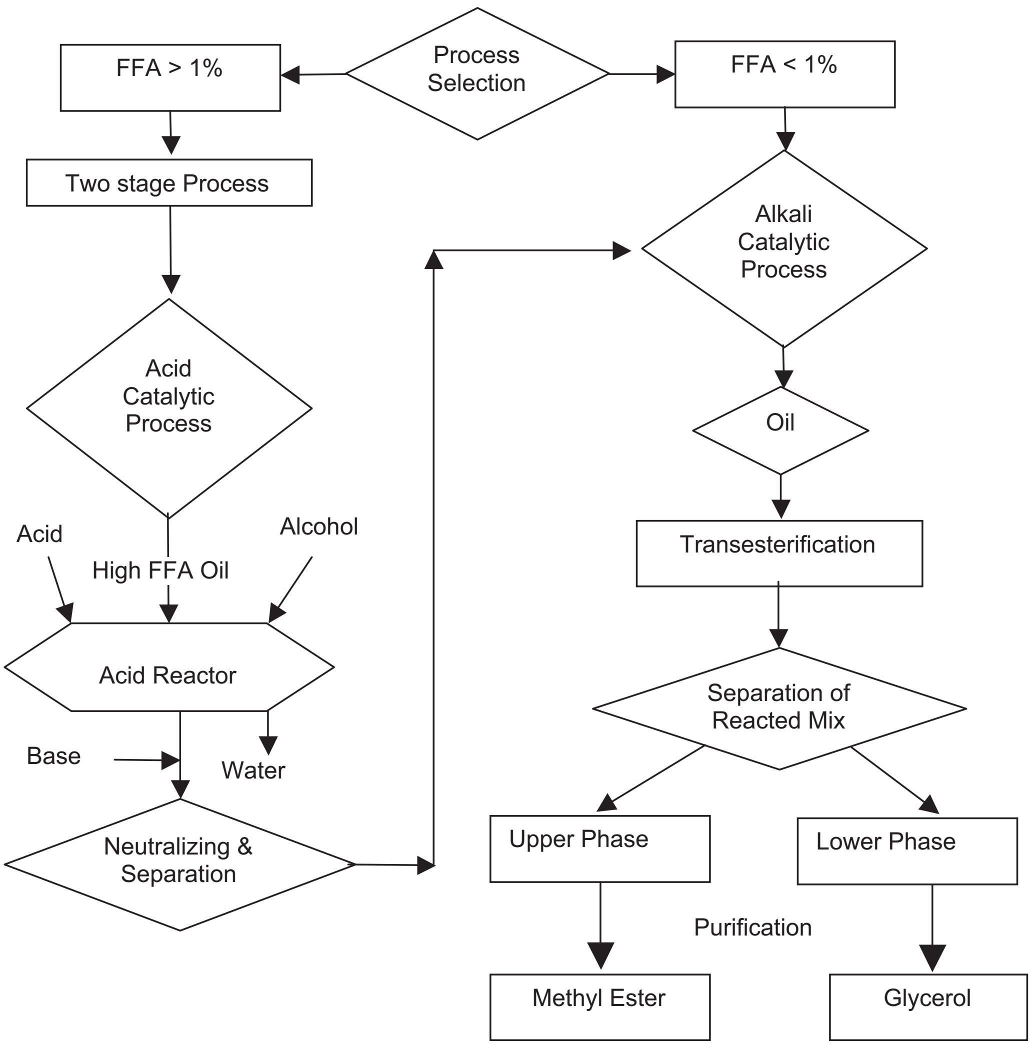

Fig. 3. Process selection and steps for biodiesel production [14].

5.6.1. Engine performance with neem biodiesel

Rao et al. [58] investigated the performance characteristics of neem methyl ester in single cylinder, four stroke, DI, water cooled Kirloskar engine (compression ratio 16.5:1, rated power 3.7 kW at 1500 rpm). They reported about a slight drop in efficiency in case of methyl ester as compared to diesel. Poor combustion characteristics of methyl ester because of its high viscosity results in drop in thermal efficiency. The brake thermal efficiency of B10, B20 blends were very close to that of diesel. Ragit et al. [47] also confirmed that BTE increases for all the blends but not more than diesel. Maximum BTE obtained with methyl ester at part load was 16.53%. But it starts decreasing with diesel at full load. BSFC of neem oil methyl ester was lower at part load and higher at full load than that of diesel [47]. BSEC of neem oil methyl ester and its blends decreased at part load and increased at full load in comparison to diesel. This is due to lower calorific value.

5.6.2. Emission characteristics of neem biodiesel

Major focus was on the emission characteristics of the neem oil methyl ester (NOME). Sundraapandian and Devaradjane [59] conducted theoretical and experimental investigation to evaluate the performance and exhaust characteristics of neat biodiesel of neem oil, Jatropha oil and karanja oil. It was found that CO, NO_x, HC and smoke emission were reduced to 18%, 3%, 18% and 12% for NOME when compared to diesel fuel. Specific gas emission based

Table 6
Process selection and operating conditions for biodiesel production from neem oil [47,48,50,51].

Transesterification process	Operating conditions				Yield (%)
	Alcohol to oil ratio	Catalyst to oil	Time	Temperature (°C)	
Alkali catalyzed					
i. Methanol	i. 6:1	NaoH – 0.7%	i. 6.5 h	i. 60	i. 94
ii. Ethanol	ii. 10:1		ii. 8 h	ii. 80	ii. 88
Alkali catalyzed methanol	0.178–0.356 (w/w)	KOH 0.01–0.03% (w/w)	0.75–1.15 h	60	–
i. Acid pretreatment for reducing FFA to 2%	i. 0.25 (w/v)	1% sulfuric acid	0.5 h	50	–
ii. Alkali catalyzed methanol	ii. 0.25 (w/v)	0.5% (w/v)	0.5 h	55	
Synthesized phosphoric acid modified catalyst	6:1	1% (w/v)		60	84

Table 7
Average changes (%) in emissions on using biodiesel blends [60,61].

Emission type	B20	B100
CO	−12	−48
Particulate matter	−2	−47
NO_x	+2	+10
SO_2	−20	−100
PAH	−13	−80

studies have been discussed in the following sections. Table 7 gives an overall idea of the changes in emission due to biodiesel use.

5.6.2.1. Nitrous oxide. The literature has reported that the engine operation on biodiesel gave lower emission than diesel fuel except in case of NO_x. Generally NO_x emission increases with increasing engine load. NO_x emission is greater for biodiesel blends than that of neat diesel fuel for the same engine load. It is reported that NO_x emission increases from 2% to 10% when B100 is used in stead of B20 [60]. Neat biodiesel contains about 10–12% oxygen in its molecule. This additional oxygen is responsible for higher NO_x emission. Emission characteristics of fresh neem oil and its blends (25%) with diesel were studied by Rao et al. [58]. They found that fresh oil showed lower NO_x emissions when compared with diesel and neem oil blends. The oil blends with diesel also showed slightly higher smoke intensities than diesel. Carbon monoxide emissions of the blends were lower compared to fresh neem oil.

With diesel–NOME blends, NO_x was reduced at retarded injection timing but increased at advanced injection timing. In the continued work, Nabi et al. [48] had investigated the combustion and exhaust emissions with diesel fuel and diesel–NOME blends (5%, 10%, and 15%). They found that exhaust emissions including smoke and CO were reduced, while NO_x emission was increased with diesel–NOME blends for all the injection timing, compared with conventional diesel fuel. It was reported that NO_x emission increases 10% in comparison to diesel when B50 blend of neem methyl ester was used [49]. Ragit et al. [47] reported that NO_x emission from a four stroke single cylinder water cooled diesel engine (compression ratio 17.5:1, capacity 661 cm^3, rated output 5.2 kW at 1500 rev/min) showed a decreasing trend when different blends of neem oil methyl ester was used at different loads and it was least at full load. The emission reduced by 3.22% at part load and 6.06% at full load. But when pure neem oil was used emission increases (41.13%) at part load and decreases (20.91%) at full load compared to diesel engine.

5.6.2.2. Carbon monoxide. Diesel–neem oil methyl ester (NOME) blends showed lower CO emissions compared with neat diesel fuel [62]. Engine CO emission reduces with increasing biodiesel percentage in fuel. CO emission increases as fuel–air ratio becomes more than stoichiometric value. It has been observed that with B50 blend, engine CO emission is reduced by 31% compared to the CO emission with neat diesel fuel [49]. But when 100% pure neem oil is used, CO emission increases because high viscosity and poor atomization tendency of neem oil lead to poor combustion [49]. Rao et al. [58] observed that CO emission increases with B60, B80 and B100 blends due to incomplete combustion. To overcome this problem they suggested a change in injection pressure and combustion chamber design.

5.6.2.3. Particulate matter. PM emission decreases with the increase in biodiesel percentages. Because of high oxygen content of biodiesel, better combustion takes place and particularly black soot burns so efficiently that there is a net reduction in PM emission. A maximum of 30% PM emission reduction is achieved with B50 blend [48].

5.6.2.4. Oxides of sulfur. Smoke is formed due to incomplete combustion. The smoke emission from engines reduces with all biodiesel blends. The smoke emission is reduced by 34% when B50 blend of neem oil methyl ester was used [48]. Ragit et al. [47] reported that smoke opacity increases at part load and decreases at full load when neem oil methyl ester is used. For pure neem oil smoke opacity increases by 70.27% at part load compared to diesel and slightly increases at full load.

5.6.2.5. Carbon dioxide. Ragit et al. [47] reported that neem oil methyl ester gives lower emission in comparison to diesel whereas neem oil gives lower CO_2 emission due to incomplete combustion. At part load neem oil gives higher CO_2 emission as compared to diesel, due to complete combustion, whereas neem oil methyl ester shows increasing trend as compared to diesel.

6. Conclusions

In developing countries like India where most of the energy demands is met by crude oil import energy security has become the main priority for the economic development of the country. As an alternative renewable energy source biodiesel from non-edible oils is becoming popular in India. Mainly Indian biodiesel mission focused on Jatropha because of its various advantages like wide adaptability, low fertilizer and irrigation requirement, pest resistance, etc. But in reality some of these perceptions have been disproved. So it has become essential to diversify the biodiesel feedstock basket for sustainable and long-term supply of biodiesel. Among other non-edible oils neem with its multiple valuable uses draws attention as a sustainable biodiesel feedstock. Neem can be used for better pest and nutrient management, reforestation, medicinal purposes, environmental purification, etc.

Though existence of non-edible oil in our country had been established long ago but there had been no systematic study, which could actually prove these to be the right candidates for biodiesel production. Neem shows great potential as a non-edible biodiesel feedstock that can be used in an integrated approach considering its multipurpose uses. In the initial phase of introduction, biodiesel is likely to be expensive than diesel. The difference between petro-diesel and bio-diesel would need to be covered by subsidy/concessions to the Oil Marketing Companies.

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